

[Supporting Information]

Highly Efficient Energy Transfer from Fluorescent Gold Nanoclusters to Organic J-aggregates

Somnath Banerjee^a, Naupada Preeyanka^a, Himani Dey^b, Sudipta Seth^c, Abdur Rahaman^b and Moloy Sarkar^{a*}

^a*School of Chemical Sciences, National Institute of Science Education and Research, Bhubaneswar, HBNI, Bhimpur-Padanpur, Jatni, Khurda-752050, Odisha, India.*

^b*School of Biological Sciences, National Institute of Science Education and Research, Bhubaneswar, HBNI, Bhimpur-Padanpur, Jatni, Khurda-752050, Odisha, India.*

^{*}*Email: msarkar@niser.ac.in*

^c*Department of chemistry, University of Hyderabad, Hyderabad – 500046, India.*

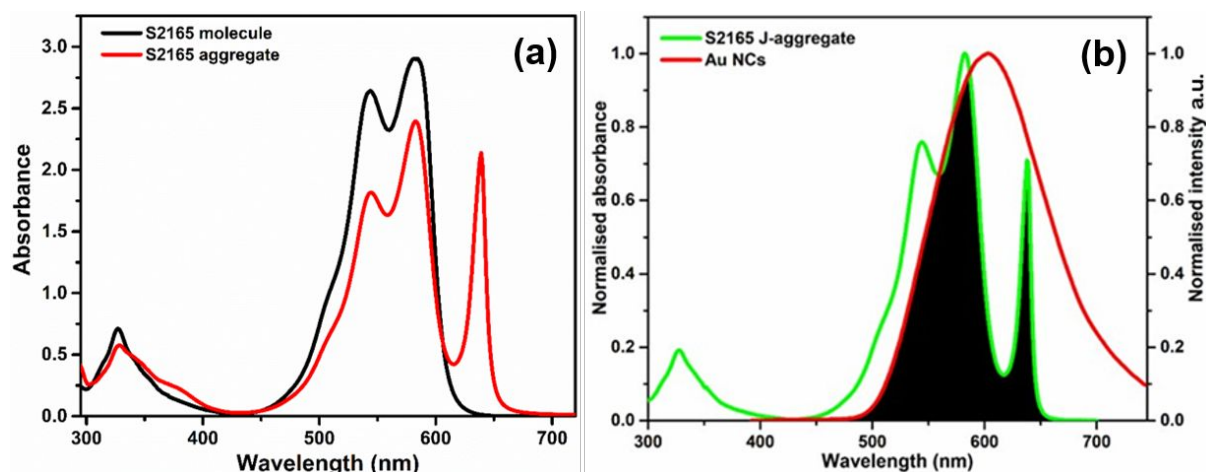


Figure S1. (a) Absorption spectra of S2165 dye in molecular (black) and aggregated form (red). (b) Spectral overlap between the emission spectrum of donor Au NCs (red) and absorption spectrum of acceptor S2165 dye aggregate (green).

Experiment in support of FRET process:

In the present study to prove that the enhancement in the emission intensity of J-aggregate (acceptor) is not due to other factors but due to energy transfer, we have performed two new experiments in support of FRET process. Please note that these experiments were well established methods used by other researchers to establish FRET or no-FRET events for different systems. Firstly, experiment is done by monitoring the fluorescence response (enhancement or no-enhancement) of acceptor in absence and presence of the donor (Au NCs).¹ Specifically, in the experiment, we have subtracted the emission coming from only J-aggregate (without Au NCs) with that of Au NCs + J-aggregate hybrid system (maintaining the appropriate concentrations on both occasions). Figure S3 (a) represent the corresponding spectra. From Figure S3 (a), it can be seen that after subtraction we still get emission corresponding to J-aggregate with almost no emission from Au NCs. This is only possible if the energy is being transferred from Au NCs (after direct excitation) to J-aggregate, otherwise we would get emission from Au NCs. Moreover, we would like to mention that the quenching of donor emission close to the absorption maximum of acceptor, change in fwhm of the

acceptor in absence and presence of Au NCs also provides support in favour of electronic interaction between donor and the acceptor moiety.²

The second experiment is done by monitoring the fluorescence response of the system by externally adding electrolytes³ keeping in mind the fact that the current FRET event is electrostatically driven and added electrolytes will modulate the donor-acceptor approach (possibly by shielding the negative charge of the S2165 dye) which eventually will modulate the FRET response. In the second experiment, we have gradually added tetraethyl ammonium perchlorate (TEAP) salt to the Au NCs + dye aggregate hybrid system and recorded the emission spectra (Figure S3 (b)). It has been found that with increase in the concentration of salt, the emission intensity of the hybrid system increases, indicating FRET process is operative in this case. As stated earlier, the FRET efficiency is dependent on the distance between donor-acceptor pair (D-A pair). Again we have plotted PL/PL_0 vs concentration of TEAP salt (Figure S3 (c)), where 'PL' and ' PL_0 ' are the emission intensity of hybrid system in presence of salt and Au NCs in presence of salt respectively. The change in the PL/PL_0 value with increase in the salt concentration also provides support in favour of electrostatic interaction between donor and the acceptor species and associated energy transfer process between them.

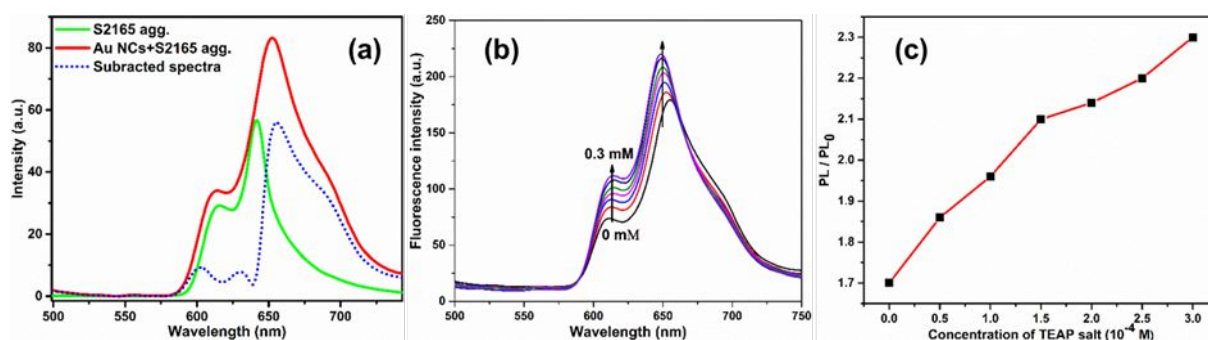


Figure S2. (a) Emission spectra of S2165 aggregate (green), Au NCs+S2165 aggregate hybrid system (red) and the subtracted spectra between them (blue dotted). (b) The emission spectra of gradual addition of tetraethyl ammonium perchlorate (TEAP) solution (0 to 0.3 mM) to the Au NCs-dye aggregate hybrid system. (c) Plot of PL/PL_0 vs TEAP concentration, showing the increase in PL intensity with the increase in concentration of the ammonium salt solution.

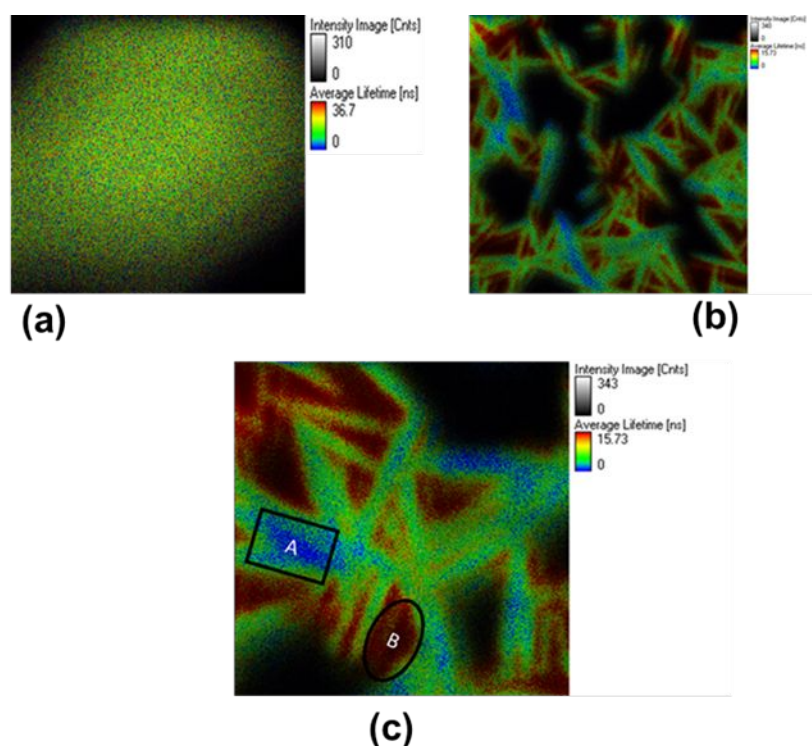


Figure S3. Fluorescence lifetime images of (a) Au NCs (total window scale bar is 5 μm x 5 μm) and (b) Au NCs/J-aggregate hybrid system recorded using excitation wavelength of 405 nm. (total window scale bar is 10 μm x 10 μm) (c) Enlarged FLIM image of Au NCs/ S2165 J-aggregate hybrid system indicating two different area (A and B) from where lifetimes have been calculated (total window scale bar is 4.6 μm x 4.6 μm).

Table S1. Growth of tetrahymena thermophila cells in absence (untreated) and presence (treated) of Au NCs and Au NCs-dye hybrid.

Time (hours)	Untreated (10^5 cells/ml)	Treated with Au NCs (10^5 cells/ml)	Treated with Au NCs + dye (10^5 cells/ml)
0	1.36	1.29	1.30
3	2.57	2.37	2.45
6	3.82	3.71	3.62
9	6.21	5.93	5.67
12	9.87	9.28	9.71

References:

- (1) Santhosh, K.; Patra, S.; Soumya, S.; Khara, D. C.; Samanta, A. Fluorescence Quenching of CdS Quantum Dots by 4-Azetidinyl-7-Nitrobenz-2-Oxa-1,3-Diazole: A Mechanistic Study *ChemPhysChem* **2011**, *12*, 2735–2741.
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- (3) He, C.; Nguyen, T. D.; Edme, K.; Cruz, M. O.; Weiss, E. A. Noncovalent Control of the Electrostatic Potential of Quantum Dots through the Formation of Interfacial Ion Pairs. *J. Am. Chem. Soc.* **2017**, *139*, 10126–10132.